Spin, Charge, and Heat Coupling at Magnetic Interfaces

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論 文 内 容 要 旨

(NO. 1)

氏 名	Adam Badra Cahaya	提出年	平成 29 年
学位論文の 題 目	Spin, Charge, and Heat Coupling at Magnetic Interfaces （磁性界面におけるスピン・電荷・熱の結合）		

Background

The magnetization dynamics in magnetic nanostructures is important for engineering of magnetic recording. In magnetic multilayers, a dynamic magnetization of magnetic medium "pumps" spin current into adjacent normal metals. The spin pumping enhances the magnetization damping, and can be interpreted as the Onsager reciprocal effect to the current-induced spin-transfer torque. Both effects are governed by the same spin-mixing conductance parameter $g^{\uparrow\downarrow}$. $g^{\uparrow\downarrow}$ also governs the spin-Seebeck effect (SSE). The physical origin of the SSE is the thermal spin pumping, which converts a heat current into a spin current. This spin current is converted into a charge current by the inverse spin-Hall effects (ISHE). SSE offers a new strategy in electric power generations and large-area applications are being considered by the Japanese electronics maker NEC. However, to be able to optimize the heat-to-spin-to-charge current conversion, the figure of merit (*ZT*) of SSE based power generators should be determined.

The $g^{\uparrow\downarrow}$ parameter depends on the interface cut and orientation to the normal metal. This anisotropy could partly be explained by the density of the local magnetic moments directly at the interface. The rotational symmetry of magnetic atoms can be broken by the electric fields generated by neighboring atoms, i.e. the so called *crystal field*. The relationship between the spin-pumping effect and the local symmetry of interface magnetic moments has, to the best of our knowledge, not been studied yet. Therefore, a local picture of the exchange interaction between conduction electrons and local moments at interfaces is needed.

Local magnetic moments in solids are formed by partially filled 3d and 4f subshells of transition metals and rare earths, respectively. The former are relatively light and their spin dynamics are dominated by the exchange interaction, with corrections by the crystal fields. Rare earths (RE), on the other hand, have their magnetic subshell shielded by outer shells, which decreases the effect of crystal-fields and allows the electrons to orbit almost freely in the central Coulomb field of the ionic core with large nuclear charges. The spin-orbit interaction (SOI) of RE is therefore large and free-atomic like. Since SOI couples the electric and magnetic degrees of freedom, we expect significant effects of electric fields on the RE magnetization dynamics that will lead to new research topic of *rare-earth interface spintronics*.

Purposes

The purposes of this thesis are:

(1) to characterize the efficiency of heat-to-charge current conversion in the spin-Seebeck effect.
(2) to determine the effect of symmetry of local magnetic moments in $g^{\uparrow\downarrow}$, and
(3) to understand the coupling between voltage and the local 4f magnetic moments at interface of ferromagnetic insulator (FI) and nonmagnetic metal (N).

Voltage generation by thermal spin pumping

Spin currents can be generated by temperature gradients across FI and N bilayer. The spin current flowing through the interface is determined by magnon temperature of FI $T^m_{\rm FI}$, electron temperature of N $T^e_{\rm N}$, and the spin accumulation μ_s at the interface on the N side. In average, the spin current is polarized along the magnetization direction. The magnitude of the net spin current is

$$J_s = -(\hbar/2e)\, G_S\, [S_S(\, T^m_{\rm FI} - T^e_{\rm N}) - \mu_s/2e], \tag{1}$$

where S_S is the spin Seebeck constant, G_S is the interface spin conductance, $\hbar$ is the Planck constant divided by 2π, and -*e* is the electron charge.

In the limit of a dominating thermal boundary resistance (Kapitza resistance), it is possible to solve the power-conversion problem analytically. The pumped spin current can be converted into charge current by ISHE or spin valve. We characterize the efficiency of such power generations. The ISHE is favorable for large area thermoelectric coatings, even though *ZT* is small. The filtering effect of spin valve structure offers the possibility to enhance *ZT* considerably.

Spin pumping by localized magnetic moments

On transition-metal and rare-earth based FI, the magnetic moments are originated from the spin and the total angular momenta of 3d and 4f electrons, respectively. In the first case, the crystal field quenches the angular momentum of 3d electrons, and locks the orientation of 3d orbitals to the crystalline directions. The spin density of conduction electrons $\mathbf{s}_c$ and local moments $\mathbf{S}_d$ are related by conservation equation

$$\partial_t \mathbf{s}_c(\mathbf{r},t) + \nabla\cdot\mathbb{J}_s(\mathbf{r},t) = J_{ex}\mathbf{s}_c(\mathbf{r},t)\times\mathbf{S}_d(\mathbf{r},t), \tag{2}$$

where $\mathbb{J}_s$ is spin-current tensor.

The right-hand side of Eq. (2) arises from the exchange interaction between localized and conduction electrons, with an exchange constant J_{ex}. The magnetic subshell acts as a spin-current source for conduction electrons, and then the deformations of the 3*d* electric cloud, characterized by a quadrupole moment Q_2, manifest as anisotropies in the $g^{\uparrow\downarrow}$.

By analyzing the spin polarization of conduction electrons, we find that the anisotropy of $g^{\uparrow\downarrow}$ depends on the angle β between crystal-field direction and interface normal (see Fig. 1). Estimation of the $g^{\uparrow\downarrow}$ for several configurations qualitatively agrees with the experimentally observed values. In the case of rare earths, the anisotropy arises from the atomic spin-orbit coupling. $g^{\uparrow\downarrow}$ becomes a function of the magnetization orientation.

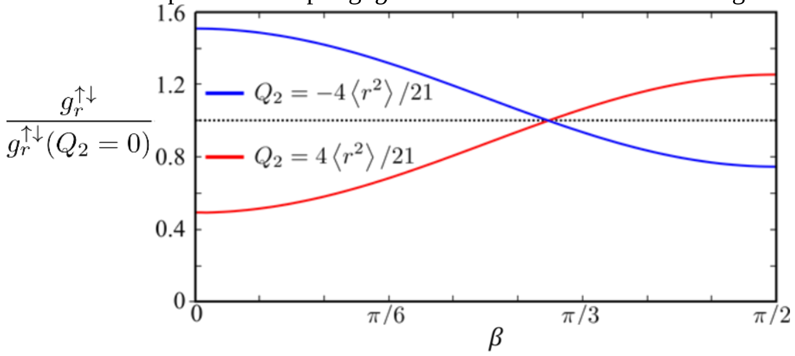

Figure 1. The real part $g_r^{\uparrow\downarrow}$ of spin-mixing conductance $g^{\uparrow\downarrow} = g_r^{\uparrow\downarrow} + ig_i^{\uparrow\downarrow}$ of a ferromagnetic insulator and a normal metal for 3d magnetic moment with quadrupole anisotropy $Q_2 = \pm 4\langle r^2\rangle/21$, as a function of the angle between crystal field direction and interface normal β.

Voltage control of magnetization by 4*f* SOI

The SOI of a 4*f* subshell strongly couples its spin and charge $[n_f(\mathbf{r})]$ densities. A 4*f* magnetic ion interacts with static electric fields, $\mathbf{E} = -\nabla\phi$, where ϕ is the electric potential. At FI|N, the electrostatic energy induces a voltage-modulated magnetic anisotropy

$$H_e = -e\int d^3\mathbf{r}\,\phi(\mathbf{r})n_f(\mathbf{r}) = H_0 - H_{ani}m_z^2, \tag{3}$$

where m_z is *z*-component of the magnetization, H_0 collects terms that does not depend on the magnetization. $H_{ani} \propto Q_2^e\Delta V$ is proportional to electric quadrupole moment, Q_2^e and voltage applied across FI|N bilayer ΔV. The voltage-induced magnetic anisotropy energy changes the ground state magnetization directions (Fig. 2b).

The coupling of magnetic and electric degree of freedom is enabled by the broken symmetry of the electric field in *z* direction due to the screening of **E** in N (Fig. 2a). At FI|N, we predict a voltage-induced ferromagnetic resonance and magnetization switching. The rare-earth-mediated torques allow power-efficient control of spintronic devices by electric fields.

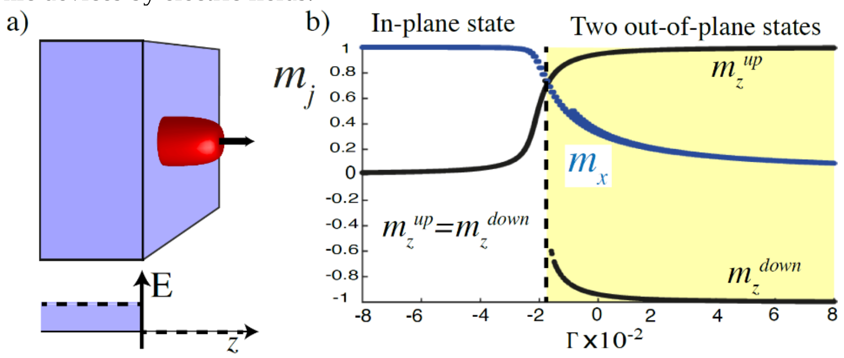

Figure 2. a) Electric field at an interface between an insulator and a metal. The magnetic dipole and charge quadrupole at the interface are strongly coupled. b) Ground state magnetization directions $\mathbf{m} = (m_x, m_y, m_z)$ as a function of interface electric field with coupling parameter $\Gamma \propto Q_2^e\Delta V$.

Conclusion

(1) The figure of merit of power generation based on thermal spin pumping depends strongly on the spin-to-charge conversion scheme. By using ISHE and spin valve as spin-to-charge conversion scheme, we offer strategies to enhance thermoelectric *ZT*.

(2) In spin pumping, the anisotropy of 3*d* and 4*f* subshells renders the pumped spin current anisotropic. The anisotropy of 3*d* and 4*f* originates from the crystal field and atomic spin-orbit coupling, respectively.

(3) The spin and charge coupling of rare earth manifests as a voltage-modulated magnetic anisotropy and enables a power-efficient control of magnetization devices by electric field.

論 文 目 次

別　紙

論文審査の結果の要旨

磁気トンネル接合において、磁気モーメントを制御する方法として、スピンにかけるトルクを磁場ではなく電流によってうみだすことが提案されている。この方法は、磁気ランダムアクセスメモリーへの応用する際、低消費電力化を実現するために重要である。最近のスピントロニクスでは、低消費電力を実現するために、（１）スピン軌道相互作用を用いたトルクの最適化（スピンオービトロニクス）や、（２）熱流を用いた、トルクや電圧の発生（スピンカロニクス）、さらに（３）電流でなく電場を用いた磁気スイッチグなどが提案されている。

Adam Badra Cahaya が提出の博士論文では、（１）スピンゼーベック効果によるスピン誘起熱流の変換効率を評価し、（２） 金属磁性体界面での局在磁気モーメントのダイナミクスをモデル化した。また（３）スピントロニクス素子において希土類元素を用いて、機能を改良することを提案している。論文は６章からなり、そのうち３，４，５章がオリジナルな仕事である。第１章では、本論文の目的と背景が述べられている。第２章では、スピンポンピングと磁気モーメントの相互作用の方法が、３ｄ電子に関しては結晶場の効果を、また４ｆ電子に関してはスピン軌道相互作用の効果をそれぞれ考慮して述べられている。第３章では、スピンゼーベック効果における変換効率の計算をしている。特にスピンホール効果によるスピン電荷変換は、大面積への応用では有用であるが、計算した効率は 10^{-4} 程度に過ぎないことを示した。第４章では、スピン輸送を、伝導電子でなく、３ｄや４ｆ電子による局在磁気モーメントで行うという新しい提案をしている。特に、金属・磁性絶縁体界面における、スピンポンピングの速度を計算し、界面における有効場とその異方性が、界面での歪やスピン軌道相互作用によることを見出している。第５章では、４ｆ電子の電気四重極モーメントが、界面に局在する磁気モーメントと相互作用することを指摘している。 この相互作用を用いれば、低消費電力でスピンの向きを変えることができることを提案している。第６章では、本論文で得られた主な結論が述べられている。

本論文で得られた結果は、スピントロニクスに新しい知見と方向を示すものである。このことは Adam Badra Cahaya が独立した研究活動を行うに足る高度の研究能力と学識を有することを示したものである。よって Adam Badra Cahaya 提出の論文は博士（理学）の学位論文として合格と認める。